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Chemical and Radioactivity Effects of Geothermal Springs on Environmental Pollution in Seferihisar Region in Western Turkey

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In this study, the toxic properties of Seferihisar geothermal springs are analyzed and the results show the negative effects on groups in the organisms environment or have a pollutative effect on the ecosystem. Some properties of Seferihisar region geothermal springs are determined as: temperature 69-67ºC, electrical conductivity (E.C.) 11200-22500 μmhos/cm, total cations 139.90-223.15 me L^{-1}, total anions 134.95-220.10 me L^{-1}, organic matter 3.30-8.10 me O_2 L^{-1}, NH_4^-–N 2.10-2.22 me L^{-1}, NO_3^-–N 1.10-1.65 me L^{-1}, NO_2^-–N 0.44-0.95 me L^{-1}, boron 11.00-16.60 me L^{-1}, COD 40-122 me L^{-1}, Fe 0.26-0.42 me L^{-1}, Cu 0.06-0.22 me L^{-1}, Pb 0.48-0.64 me L^{-1}, Zn 0.06-0.13 me L^{-1}, Cd 0.040-0.090 me L^{-1}, Cr 0.04-0.13 me L^{-1}, Co 0.09-0.18 me L^{-1}, Si 33.60-71.30 me L^{-1}, ^{222}Rn 13.79-229.89 pCi L^{-1} and ^{226}Ra 2.1-24.52 pCi L^{-1}.

Key Words: Radioactivity, Geothermal Springs, Pollution, Boron, Radon.

INTRODUCTION

All living creatures showing dispersion in nature live in harmony and balance both among themselves and by interacting with other elements of the environment. The balance that the creatures have with their environments gives them a healthy aspect. The fact that this balance is disturbed from time to time causes formal changes in the ecological appearance.

One of the most intensive effects on soil, air and water pollution is the geothermal springs. Waters coming out of geothermal springs form part of the underground water that comes to the surface and these are identical to the temperatures of the underground water from where they originated. Waters of geothermal springs are classified as magmatic (juvenile) or fossil (reservoir system) by the origin of the fluid in the reservoir rock. Hot waters come out from depths of the earth where there are fault lines and they are the origins of the magma-contacted juvenile waters so that they form the origin of geothermal springs. Due to their excellent resolving

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properties, geothermal waters, contain soluble salts and heavy metals depending on the chemical contents of the rocks. They pollute the rivers, underground waters and soils in the vicinity and turn them into an unusable form. In polluted waters, photosynthesis and respiration processes are disturbed and so does the balance of ions.

The Seferihisar geothermal springs is one of the important geothermal areas in western Anatolia of Turkey. Geothermal activities of the area have been known since ancient times, tracing back to the Roman period. People once used the thermal waters in this area for bathing and washing purposes. Thermal waters formed travertine walls as they flowed along ancient aqueducts. These waters are presently used for bathing and primitive spa facilities. The Seferihisar geothermal field and its surrounding are an important touristic resort due to the historic sites, beaches and tangerine gardens. The study area is located 70 km southwest of Izmir city center (Fig. 1). There are five natural thermal spring groups in the area. The thermal springs have total discharge rates of 100-150 L/s and their temperatures vary from 30 to 78ºC. Twenty geothermal gradient wells having depths of 56-171 m and ten deep geothermal wells having depths of 151-2000 m have been drilled by MTA (General Directorate of Mineral Research and Exploration of Turkey). Unfortunately, these thermal wells and springs are not presently used except for primitive spa facilities. Additionally, they can cause some environmental problems for groundwaters and surface waters as they flow uselessly and because their salinities and boron contents are high and harmful for the tangerine gardens in the area. The district heating project of some residences of Izmir city and Seferihisar town use the Seferihisar geothermal waters, as planned by the Izmir Governmental organization. Electricity generation, greenhouse heating, balneological, thermal tourism and heating of swimming pools are the other uses which can be developed for this area. Several geothermal surveys were carried out in this and surrounding areas. This research discusses the hydrogeochemical characteristics and radioactivity levels of the thermal springs in detail for the study area. Special emphasis is put on the study of geothermal springs effects on environmental pollution.

Some of the studies on environmental pollution of thermal waters relate to radioactivity. Natural radionuclides present in environment can be found in variable proportions in thermal waters. The most commonly found radionuclides belong to the uranium and thorium family present in groundwater. High concentrations of natural radioactive elements found in thermal waters. These concentrations depend on the physico-chemical conditions and on the geological environment[1].

The radioactivity effect of thermal waters is due to radon. Radon is an inert gas generated from radioactive decay of the uranium decay chain and is the only natural gaseous radioactive tracer. These gases are poorly affected by chemical equilibration processes.

The greatest source of radon in an underground facility is the radon which escapes from ground water. As a rule, the radon concentration which occurs in the intramontaneous groundwater is 100-1000 Bq L^{-1} (2702.7-27027.0 pCi L^{-1}) with maximum concentration of 20.000-50.000 Bq L^{-1} (540540-1351350 pCi L^{-1}) in water arising from uranium-rich rocks. The reason for the high concentration of radon in ground water is that six-valence uranium is relatively easily soluble and is leached out of the rock by the groundwater[2].

In the study, it has been found that the radon concentrations in thermal waters in Western Anatolia ranged from 0.14 Bq L^{-1} (3.78 pCi L^{-1}) to 5.77 Bq L^{-1} (155.95 pCi L^{-1}), averaging 1.45 Bq L^{-1} (39.19 pCi L^{-1})[3].

In other study in Northeastern Tunisia, it has been found that the radio-activities in thermo-mineral sources ranged between 0.034 Bq L^{-1} (0.92 pCi L^{-1}) and 3.9 Bq L^{-1} (105.41 pCi L^{-1}) for ^{226}Ra.

Singh *et al.*[2] emphasize that changes in radon concentration in ground water differ significantly with time. They found that the average value of radon concentarion from daily monitoring is 0.71 (19.19 pCi L^{-1}) Bq L^{-1} with a standard deviation of 0.84. According to results, they clearly observed that there was a seasonal variation in radon level which tends to be high at the end of the summer season and low at the of the winter season.

EXPERIMENTAL

Water used in this study were sampled from geothermal springs near the township of Cumali, Tuzla and from Karakoç stream in which both spring waters and surface waters using two litre of bottles. Samples were collected in year 2004-2005 season for the investigation and for effects on environmental polution of the thermal waters.

Geological properties

In accordance with the lithological and hydrogeological properties of geological units in the study area are divided into four main groups (Fig. 1). These units are: (1) Upper Cretaceous-Paleocene Bornova melange; (2) Neogene terrestrial sediments; (3) Neogene rhyolitic volcanics and (4) Quaternary alluvium. The basement rock of the regional geology is Precambrian to Paleocene Menderes Massif metamorphic rocks[4], which is made up of schists and marbles, and located about 10 km east of the mapped area (Fig. 1).

Holocene travertine and saltpan formations are also observed in and around the thermal springs. As with many other geothermal systems in western Turkey, the circulation of the thermal waters in this geothermal field is also closely related to major fault and fracture zones. Major faults,

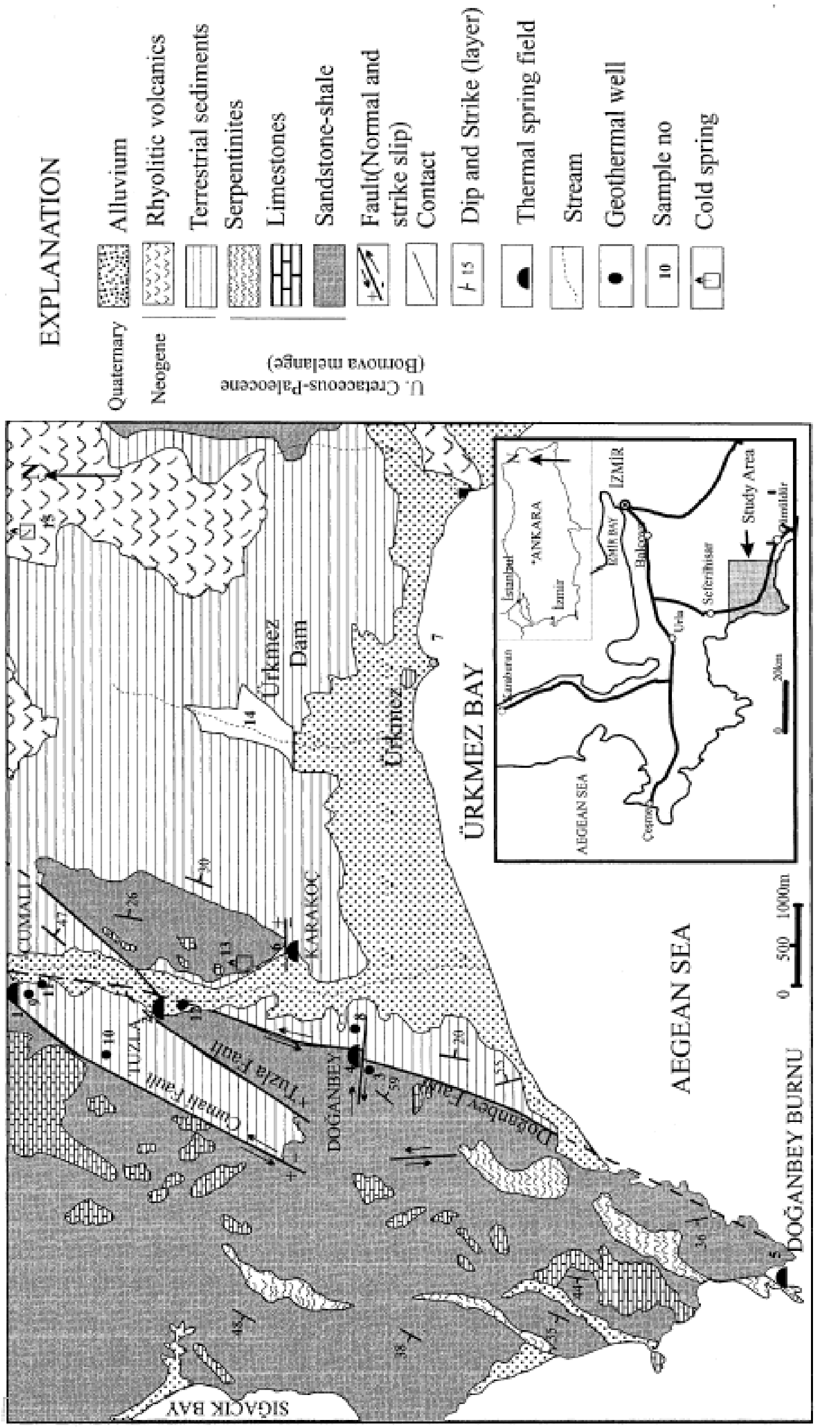

Fig. 1. Geologic map of the Seferihisar geothermal area and locations of the geothermal fields and sampled waters[6]

which are called the Cumali, Tuzla and Doganbey faults, exert structural control on the geothermal systems of the area. The area between the Cumali and Tuzla faults suggests graben structures. In this graben the thickness of the Neogene sediments attains 400 m. The Cumali fault was identified as a thrust fault by some authors[5].

The elements were analyzed and determined as calculated evaporation residue[7], pH[8], electrical conductivity[7], amount of Na^+, K^+, Ca^{2+} solved in water, flame spectrophotometer, Ca^{2+}+Mg^{2+}, Cl^- in titration[7], CO_3^{2-}, HCO_3^- in titration, SO_4^{2-} gravimetric[8], Boron carmine indicator[7], phosphor[8], NO_3^- –N colourometric[9], NO_2^-–N[10], NH_4^+–N[11], SAR, residue Na_2CO_3 and quality of irrigation waters[7], need for chemical oxygen[12]. Elements of Fe, Cu, Pb, Zn, Cd, Cr, Co, Mn, Ni, Si, and Al in water samples in an atomic absorption spectrophotometer[13].

Radioactivity measurements in thermal waters

The water samples were taken from the sources into 100 mL of radon bubblier bottles, the tops of the bottles were sealed to prevent radon leaking from the water and kept to be analyzed for the following day. The radon concentrations in thermal waters were depicted by using collector chamber method used in studies[14] (Fig. 2). In this method, the radon gas in 100 mL of bottle sucked into a vacuumed chamber after foamed by air. The products, produced by decomposing radon gas, collected on the collector plate by applying a voltage between the chamber and the collector plate. Radon in waters was measured by using a copper plate with a diameter of 4.5 cm, a collector chamber having a volume of 2.47 L and voltage of 600 V. This conditions were optimal for the efficient determination of ^{222}Rn in environmental samples by the collector chamber method[14].

After de-emanation into the chamber, the samples were allowed to stand for at least 4 h under a high electrostatic field. The α-disintegration is directly measured by the α-scintillation counter system (Eberline Model SAC-4). The counting time was 50 min for each sample. The efficiency of the collector system for water samples was determined as 12 % using ^{226}Ra standard solution with different activities. For radium activity measurement, the outlet of a compressed gas regulator (aged air) was connected to the inlet of the bubbler and gas bubbled through the water for 20-30 min. The primary purpose of this gas is to replace the air in the bubbler with the purge gas in order to remove essentially all ^{222}Rn from the water samples. De-emanation was performed at least 7 d after purging. For the water samples, most of the tests were carried out with solutions containing several Bq of ^{226}Ra. Calibration solutions, for each of 100 mL volumes, were prepared by adding distilled water and HCl to 10 mL of standard ^{226}Ra solutions, containing activities of 0.185, 0.370, 0.555, 0.740, 0.925 Bq. The efficiency of the collector system for water samples was found 12 %.

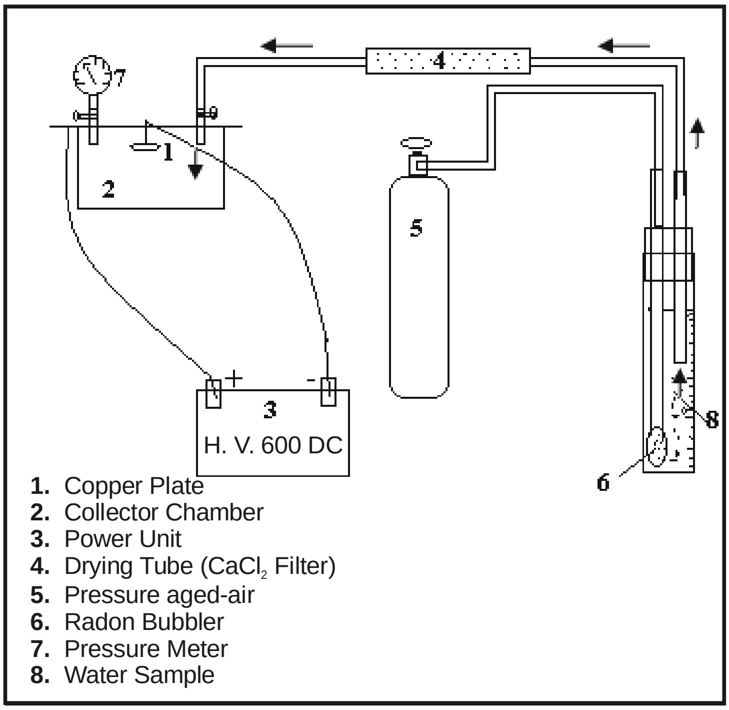

Fig. 2. Schematic diagram of radon measuring device

RESULTS AND DISCUSSION

Chemical and physical properties of geothermal spring waters

The region where geothermal springs of Seferihisar originate is geologically a limestone formation. Physically, the waters are not turbid and are odourless. They are clear but with a trace of salt. Temperature in three different springs varied from 64-79ºC (Table-1).

pH of the spring waters were 6.50-6.71 and considering they are neutral with slight amount of base. Electrical conductivity was between 11200-22500 mmhos/cm. Due to this property, they were placed in the over salinity class-6. For this reason they can not be used as irrigation water in agriculture

According to another study, it was suggested that the hypersaline thermal water of Tuzla is derived from dissolution of salt deposits. In his study, the high Na (20,000 me/L), Ca (3000 me/L), K (2000 me/L) and B (29 me/L) concentrations reflect the mineralogical composition of these deposits with a possible mineral assemblage of gypsum and Ca- and Na-borates. This mineral composition is typical for many Neogene salt-deposits in western Turkey[15].

TABLE-1
RESULTS OF PHYSICAL AND CHEMICAL ANALYSIS OF THE GEOTHERMAL SPRING WATERS OF SEFERIHISAR REGION

Places of Sampling	Karakoç	Tuzla	Cumali
Temperature of waters (ºC)	64	75	71
pH	6.56	6.71	6.50
EC × 10^6	11200	12000	22500
Cations (me L^{-1})			
Ca^{2+}	21.00	24.00	32.00
Me^{2+}	6.10	4.60	5.80
Na^+	102.20	109.50	180.50
K^+	3.60	2.95	4.85
Total of Cations (me L^{-1})	132.90	141.05	223.15
Anions (me L^{-1})			
Cl^-	94.50	103.22	140.60
SO_4^{2-}	19.80	27.42	44.50
HCO_3^-	20.60	14.00	35.00
CO_3^{2-}	–	–	–
Total of Anions (me L^{-1})	134.95	144.64	220.10
SSP	76.89	77.63	80.88
SAR	27.77	37.31	41.58
Bor (me L^{-1})	11.00	12.90	16.60
Evaporation residue (g L^{-1})	9.60	14.50	18.10
Water quality of irrigation	C_4 - S_4	C_4 - S_4	C_4 - S_4
Phosphor (me L^{-1})	0.045	0.056	0.060
Organic material MeO_2/L	3.80	4.80	8.10
COD (me L^{-1})	40	82	122
NH_4^+– N (me L^{-1})	2.22	2.45	2.10
NO_3^-–N (me L^{-1})	1.65	1.40	1.10
NO_2^-–N (me L^{-1})	0.44	0.64	0.95
Fe (me L^{-1})	0.32	0.26	0.42
Cu (me L^{-1})	0.06	0.07	0.22
Pb (me L^{-1})	0.48	0.51	0.64
Zn (me L^{-1})	0.06	0.11	0.13
Cd (me L^{-1})	0.04	0.06	0.09
Cr (me L^{-1})	0.04	0.04	0.13
Co (me L^{-1})	0.09	0.16	0.18
Mn (me L^{-1})	0.03	0.04	0.04
Mo (me L^{-1})	trace	trace	Trace
Ni (me L^{-1})	trace	trace	Trace
Si (me L^{-1})	33.60	59.40	71.30
Al (me L^{-1})	trace	trace	Trace
Radionuclides (average pCi/L^{-1})			
^{222}Rn	84.47	62.02	103.08
^{226}Ra	7.41	6.73	11.02

In terms of cations causing the electrical conductivity to intensify and anions with identical contents, Ca^{2+} was analyzed with limits of 21.00-32.00 me L^{-1}, Na^{+} 102.20-180.50 me L^{-1}, K^{+} 2.95-4.85 me L^{-1}, Mg 4.60-2.10 me L^{-1}, Cl^{-} 94.50-140.60 me L^{-1}, SO_4^{2-} 19.80-44.50 me L^{-1}, HCO_3^{-} 14.00-35.00 me L^{-1}, while CO_3^{2-} was not observed. According to this data, cation distribution followed the sequence of Na^{+} >, Ca^{2+} >, Mg^{2+} >, K^{+}, anions Cl^{-} >, SO_4^{2-} >, HCO_3^{-} >, CO_3^{2-} and the dominant salt is NaCl. In the similar study discussed that sodium–HCO_3^{-} waters are common in geothermal systems associated with metamorphic rocks which is consistent with the general lithology of the Menderes Massif and the high CO_2 content that is typical of the thermal water of western Turkey[16].

$CaCO_3$ as residues were found in high concentrations in areas where geothermal springs appear. Meanwhile, Na/K percentages were found as high level and this suggests that the temperature in the reservoir rocks from thermal springs come out are high.

These waters were placed in the category of very high sodium content because they were analyzed with limits of ratio of sodium as compare to other cations (SAR) being 27.77-41.58 and soluble sodium percentage (SSP) being 76.89-80.88.

Since the temperature of geothermal springs is high and also good solvent properties, the residue of evaporation depends on the total amount of the material solved in the water between 9.60-18.10 %. HCO_3^{-} and SO_4^{2-} are also present when NaCl exist which constitutes the residue of evaporation. According to these results, we may propose that thermal waters try to balance themselves in terms of chemical ions. This balance is together with higher temperatures.

Boron contents was found at a high level (between 9.90-10.20 me L^{-1}). While > 3.75 is an unbearable limit for boron-resistant plants, data obtained from the region is three times higher than this. Although boron minerals dissolve difficult, they become concentrated due to some minerals which are solved and carried out by thermal waters while passing through formations of rock in the region. Geothermal springs in this region are juvenile waters because they come from magma and contain geothermal energy.

These springs come out as hot vapour from different depths along the fault line stretching southward between Izmir and Urla cities. These come to the land surface from regions where conglomerates, sandstone, marl with clay.

In fact, geothermal waters are natural springs coming up to the surface from the depths of the earth containing dissolved metal salts with different concentrations. These springs in the region are observed between horst and graben on faults. Their phosphorus content is very low, being at the limit of 0.045-0.060 me L^{-1}. A great deal of the phosphor in domestic waste results from food wastes, the rest coming from synthetic detergents and this has been determined as 0.85-6.00 me L^{-1} in Izmir Meles Stream (Yesildere)[17].

Organic matter contents is at the limit of 3.80-8.10 mgO_2 L^{-1} and this is fairly concentrated. According to this data they show a distribution over the limit of 3.5 mgO_2 L^{-1} accepted by TSE (Turkish Standards Institute)[18]. The necessity for chemical oxygen changes between 40-122 me L^{-1} and according to this criteria, some of them have been observed to be over the permitted limit of 70 $mgOL^{-1}$. Depending on the concentration of organic matter, a continuous increase is at stake concerning the necessity for chemical oxygen. In this context, COD data can be accepted as the criterion for organic matter.

The organic nitrogen forms in geothermal springs are between the following limits; NH_4^-–N 2.10-2.45 (me L^{-1}), NO_3^-–N 1.10-1.65 (me L^{-1}), NO_2^-–N 0.44-0.95 (me L^{-1}) and were determined with rather low values. TSE criteria[18] describes water unclean if its content of NO_3^-–N is over the measure of 45 me L^{-1}, so these results suggest that these waters are not unclean in terms of their nitrogen contents. These properties naturally show that thermal waters come from depths of the earth and that organic matter and nitrogen, which constitutes 5 % of the organic content, were not found there, that nitrogen is present in the organic structure of the organic compounds, and has not been mineralised yet. The thermal temperature that causes a decrease in the amount of nitrogen is over 150ºC[19]. But in the waters of Melez stream (Izmir) in which domestic waste is mixed in, NH_4^+–N and NO_3^-–N were analysed in high values 2.70-28.80 (me L^{-1}), 83.70-120.90 (me L^{-1}), respectively[20].

Data obtained from the analysis shows that the basic salt in the thermal waters is NaCl, which is alkali-chlorine. Generally the salts thermal waters contain are related to their depths and so in geothermal waters coming from depths up to 500-700 m SO_4^{2-} is dominant, at 700 m, HCO_3^- is dominant and, on the other hand, Cl^- anion is dominant in very hot waters. Thus, the waters which were investigated in present studies show that they came from depths of 700 m and more due to the concentrations of Cl^- ions in their content. Mean while, a high percentage of B, Cl^- and Na^+/K^+ shows that the temperature in the reservoir of the thermal waters might be high[19]. In geothermal systems, elements such as chlorine and boron dissolve from rocks and minerals are combined with the thermal waters. Especially in hot waters, as a result of the reactions of rocks under the earth's crust, chlorine ions reach at thousands of mg L^{-1}, while boron to reaches the limits of 20-30 me L^{-1}. The ratio of Cl^-/SO_4^{2-} rises and pH is medium acid-slightly alkali. CO_2, H_2S and NH_3 are the broken gasses.

Heavy metal distributions were determined as Fe 0.26-0.42 (mg L^{-1}), Cu 0.06-0.22 (mg L^{-1}), Pb 0.48-0.64 (mg L^{-1}), Zn 0.06-0.13 (mg L^{-1}), Cd 0.04-0.09 (mg L^{-1}), Cr 0.04-0.13 (mg L^{-1}), Co 0.09-0.18 (mg L^{-1}), Mn 0.03-0.04 (mg L^{-1}), Mo trace, Ni trace, Si 33.60-71.30 (mg L^{-1}), Al trace.

Temperatures of the waters of Karakoç stream, which carries other thermal waters into the sea and into which surface waters are mixed, was found to be 26ºC and pH (7.49) to be slightly alkaline, electrical conductivity (10200 μmhos/cm) to be class-6, Boron (9.00 mg L^{-1}), Chlorine (113.08 me L^{-}), sodium (126.08 me L^{-1}) were found to be high in concentration, and very high according in and SSP (55.10) and SAR (19.10) data. While it has been observed that these waters were polluted by geothermal sources and that this pollution has been carried into the sea, laboratory findings suggest that they could be used for agricultural irrigation.

The Na-Cl thermal waters of Seferihisar have measured temperatures of 30-153ºC. The heat source is the high geothermal gradient caused by graben tectonics of the area. Na^+ and Cl^- ions mainly dominate the chemistry of thermal waters, thus thermal waters of the Seferihisar area appear to be mixtures of groundwaters and seawater. The seawater contribution in thermal waters varies from 10 to 80 %[6].

In the other study, it has been reported that the physical and chemical properties of thermal water are due to mechanisms of water-rock interaction. In the study, it has been observed that good correlations between different dissolved salts and temperature indicate that the chemical composition of the thermal waters from non-marine geothermal systems is controlled by: (1) temperature dependent water-rock interactions; (2) intensification of reactions due to high dissolved CO_2 and possibly HCl gasses; and (3) mixing with overlying cold groundwater[21].

In other study, some properties of Içmeler (Seferihisar) region geothermal springs[22] are determined as; temperature 69-67ºC, electrical conductivity (E.C.) 8800-11000 μmhos/cm, total cations 133.08-171.77 mg L^{-1}, organic matter 4.44-4.86 mg O_2 L^{-1}, Boron 9.90-10.20 mg L^{-1}.

Radioactivity levels of geothermal waters

The radon concentrations obtained by monthly measurements in thermal waters varied from 13.79 pCi L^{-1} to 229.89 pCi L^{-1} with an average value of 83.19. pCi L^{-1}. In addition, the radium activities were found to be between 2.1 pCi L^{-1} and 24.52 pCi L^{-1} with an average value of 8.39 pCi L^{-1}. The maximum radionuclide concentrations were found in groundwaters of thermal spring at Cumali. Experimental data for the period from November 2004 to December 2005 for the three selected stations are shown in (Fig. 3).

It was observed that the radon and radium results increased at the ends of the winter and summer seasons.

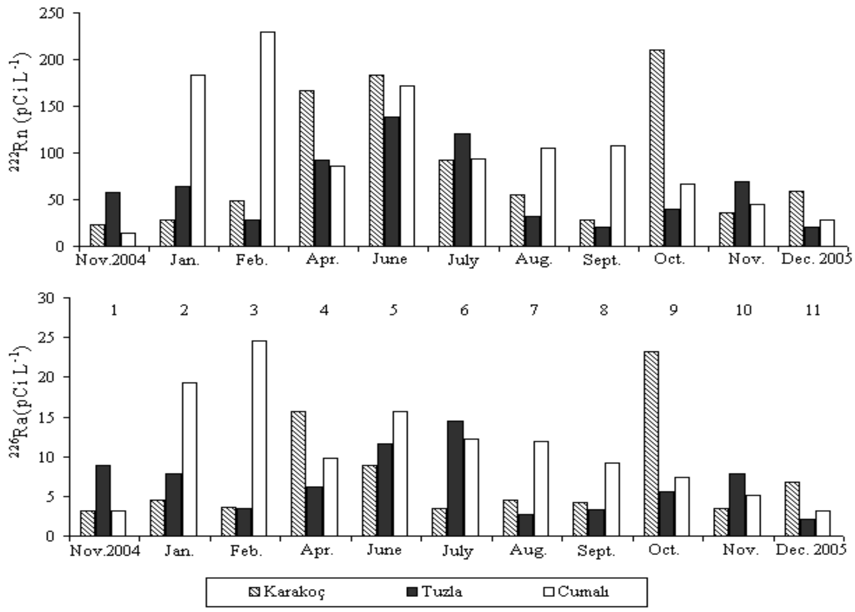

Fig. 3. Seasonal variation of radon and radium concentrations in thermal waters

Conclusion

With increasing concentration due to the lithology of the region, depth and temperature of the thermal waters, elements originating from thermal springs show dispersion on the surface that presents a waste forming medium and leaks into Karakoç stream. As a result, they act as a polluting agent. Ions which geothermal springs contain and the gases present in the vapour phase such as CO_2, H_2S, CH_4, NH_2, *etc.*, caused intense chemical precipitation, accelerating the pollution process in the region.

The radon and radium concentrations of thermal waters change significantly by time. These changes are not simple and contain inconsistent features. We observed irregularities in the radon data. There is a seasonal variation in radon level which tends to be high at the end of the summer season and low at the end of the winter season. The anomalous of the radon data are assumed to be associated with the seismic processes preceding and accompanying the earthquake events in region. Then, it has been observed to be high at October, 2005. It was occurred 4 seismic activities from M = 5 to M = 5.9 during October month[23].

Thus, the high values of physical and chemical characteristics are due to mechanisms of water-rock interaction and effect significantly surrounding of thermal source.

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